Neutron Induced Radioactivity in Certain Rare-Earth Elements

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By activating specimens of lanthanum, neodymium, samarium, europium, gadolinium, and terbium in the Oak Ridge pile for a particularly long exposure, several hitherto unobserved radioactivities are found. Determinations are made of the half-lives and of the beta- and gamma-energies by absorption and spectrometric methods. Lanthanum and neodymium were the only elements showing no internal conversion lines. Terbium appears as a contaminant in its neighboring elements by virtue of its very large cross section for neutron capture. Terbium and europium show complex beta-spectra, and each has many conversion electron lines.

I. INTRODUCTION

OXIDE specimens of the elements; lanthanum, neodymium, samarium, europium, gadolinium, and terbium were irradiated in the Oak Ridge pile for exposures of 2 months' duration. These are the lighter rare-earth elements with atomic numbers ranging from 57 to 65. The reaction expected in the pile is usually the neutron capture process, but it also appears possible to produce the excited state of stable isotopes where such long-lived activities exist. Short-lived activities are not observed in this investigation as observational study did not begin until three days after the end of the bombardments. By virtue of the very long intense neutron bombardment, many activities not previously observed have been found to exist.

Each specimen was divided into parts, for its half-life determination and for the evaluation of beta- and gamma-energies by absorption methods and by magnetic spectrometers. The results obtained with europium and with terbium have been, in part, reported[1] previously.

II. TERBIUM

Terbium exists in nature as a single isotope of mass 159. By neutron capture the radioactive isotope of mass 160 should be made. Previous studies[2–4] had assigned half-lives of 3.9 hours and 72 days to this isomeric activity. The longer half-life is in this investigation found to be 77.3 days. The gamma-radiation emitted is found by the photographic spectrometer to give, in all, about nine electron lines by internal conversion. These lines are shown collectively in Fig. 1 along with the K–L–M–N energy levels in the next heavier element dysprosium, plotted to the same scale. It is thus apparent that certain groups of the observed lines combine to represent a single gamma-ray. The energies of the observed lines together with the interpretation and the energy of the resulting four gamma-rays are shown in Table I.

In one combination a line due to converted 'N' electrons is observed for the first time. This portion of the photographic plate is shown enlarged in Fig. 2. The ratio of the L, M, and N conversion intensities can be judged from this photogram.

By absorption methods a gamma-ray of energy 1.15 Mev is found to be present. There is evidence of the existence of other gamma-rays of intermediate energy which are only slightly converted.

The beta-spectrum of terbium is found to be complex, that is, there are two overlapping spectra whose upper limits of energy are 546 and 882 kev, respectively. This activity was studied on a thick electron lens spectrometer. The observed electron distribution with momentum was converted so as to represent graphically the Fermi function *versus* energy as shown in Fig. 3. It is apparent that at higher energy a very straight line curve is obtained except for some photoelectrons due to the energetic gamma-radiation. By the proper subtraction of this projected curve at lower energies a second

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[4] G. Hevesy and G. Levi, Nature **137**, 185 (1936).

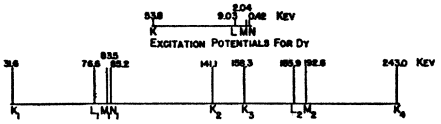

FIG. 1. Energy of conversion lines from terbium (160).

FIG. 2. Enlarged low energy spectrum of terbium (160) showing an 'N' electron group.

Fermi curve is obtained as shown, whose intercept with the energy axis is not quite as exact as that of the high energy intercept.

Terbium is of interest since it has a very large cross section for neutron capture. If present in only a slight amount as an impurity in another element its spectrum is disturbingly prominent.

III. LANTHANUM

Lanthanum consists of a single stable isotope of mass 139. By neutron capture it converts to radioactive lanthanum of mass 140. Previous investigations[5] had ascribed to this isotope a half-life of 40 hours with the emission of beta-radiation and gamma-radiation each characterized by several distinct energies. The analysis of the decay curve of this strongly irradiated sample shows clearly two half-lives, namely 43.4 hours and another about three years, not yet accurately determined. In the photographic spectrometer no conversion lines due to low energy gamma-radiation were observed in a reasonable exposure of five days.

A strong gamma-ray of 1.6 Mev was observed in connection with the shorter half-life by absorption methods. In the long-lived activity a gamma-ray of 0.79 Mev was found by absorption, and an upper limit of the beta-spectrum of approximately 0.9 Mev.

It is possible that the longer half-life here observed is really in an excited state of cerium 140 produced by beta-emission from lanthanum 140. This activity had previously been reported as having a half-life of 140 days.

IV. NEODYMIUM

Neodymium has several stable isotopes ranging in mass from 142 to 150. Previous investigations[6] had indicated the existence of radioactive isotopes of mass 141 and 147 whose half-lives were 2.5 hours and 47 hours, respectively. The present investigation shows the presence of three half-lives, whose values are 50 hours, 11.8 days, and 1.7 years. Absorption methods reveal a gamma-ray of energy 0.72 Mev and by the Feather rule beta-radiation with an upper limit of 0.74 Mev. This radiation seems to be associated with the very long-lived activity. No conversion electrons due to gamma-radiation could be observed in the spectrometers.

V. SAMARIUM

Several stable isotopes of samarium with masses ranging from 144 to 154 exist in nature. Samarium 148 exhibits natural radioactivity with a half-life of 1.6×10^{11} years. Previous researches[6,7] had revealed an induced radioactivity of 47-hour half-life ascribed to isotope of mass 153, and other activities in samarium derived from fission ascribed to isotopic masses greater than 154.

An exposure of the irradiated samarium in the beta-ray spectrometer showed several electron lines due to gamma-conversion. These lines are shown collectively in Fig. 4. One group of such lines had energy differences identical with the differences in the levels characteristic of the next heavier element, europium, as expected if the gamma-emission follows beta-radiation. The en-

TABLE I. Conversion electron spectrum of terbium.

Observed energy	Identification	Gamma-energy
31.6 kev	K_1	85.4 kev
76.6	L_1	85.6
83.5	M_1	85.5
85.2	N_1	85.6
141.1	K_2	194.9
158.3	K_3	212.1
185.9	L_2	194.9
192.6	M_2	194.6
242.7	K_4	296.5

[5] P. Weimar, M. Poole, and J. Kurbatov, Phys. Rev. **63**, 67 (1943); R. Osborne and W. Peacock, Phys. Rev. **69**, 679 (1946); C. Mandeville and W. Scherb, Phys. Rev. **73**, 1437 (1948).

[6] J. Kurbatov, D. MacDonald, M. Poole, and L. Quill, Phys. Rev. **61**, 106 (1942); C. S. Wu and E. Segré, Phys. Rev. **61**, 203 (1942).

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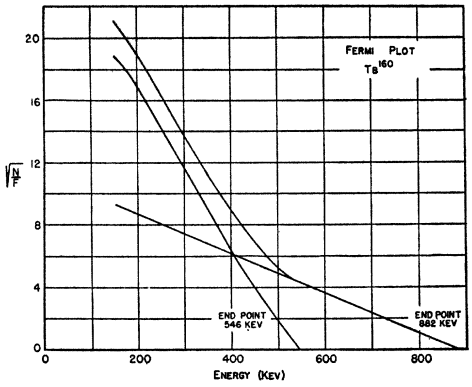

FIG. 3. Fermi plot of the beta-radiation from terbium (160).

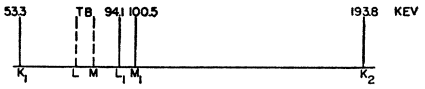

FIG. 4. Energy of conversion electrons from samarium, immediately after bombardment.

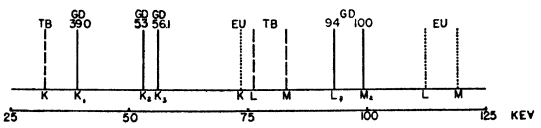

FIG. 5. Energy of conversion electrons observed from a gadolinium source, showing europium and terbium impurities.

ergy of this gamma-ray is 102.1 kev and it was found to die out with the half-life of 50 hours.

It was noted that certain of the lines agreed exactly with those observed in radioactive terbium, thus indicating the presence of this element as an impurity in the samarium. One additional line interpreted as a '*K*' conversion line indicates a gamma-energy of 242.4 kev in samarium associated with a long half-life.

The decay curve of samarium showed in addition to the 50-hour half-life and the 77.3-day terbium period, some activity of half-life greater than 150 days.

VI. GADOLINIUM

Several stable isotopes of gadolinium exist in nature. The strongly irradiated specimen of supposedly chemically pure gadolinium, yielded a spectrogram of many conversion lines in the magnetic spectrometer. These lines, 12 in number, are shown graphically in Fig. 5. From the information gained in the study of other elements it was at once apparent that both terbium and europium existed as impurities in the gadolinium. To show the ease of identifying such impurities the spectra of the three elements are shown together in Fig. 6. On subtracting the known lines due to terbium and europium, it is presumed that those that remain are really due to gadolinium. Peculiarly, however, one group of three lines has a *K–L–M* relationship exactly like that observed for the 50-hour samarium activity of energy 102 kev yet here the half-life is long. This is as would be expected if the activity in

TABLE II. Collected data on the radioactivity of the rare-earth elements.

Atomic number	Element	Half-life	Beta-radiation (Mev)	Gamma-radiation: Absorption (Mev)	Gamma-radiation: Spectrometer (kev)	Probable active isotope
57	Lanthanum	43.4 hr.		1.60		140
		~3 yr.	.90	0.79		140
60	Neodymium	50 hr.				
		11.8 da.		0.72		147
		1.7 yr.	0.74			
62	Samarium	50 hr.	1.23		102.1	153
		>150 da.		.95	242.4	155
63	Europium	~6 yr.	0.751, 1.57	1.23	39.9, 122.1, 247.3, 286.0, 342.8, 407.8	154 or 152
64	Gadolinium	76 hr.		2.10		
		—			102	153
		Long			91.0, 108.1	159 or 161
65	Terbium	77.3 da.	0.546, 0.882	1.15	85.6, 194.8, 212.1, 296.5	160

gadolinium is positron emission from mass 153, and the short-lived samarium is due to electron emission from samarium of mass 153, both transitions ending in the same excited levels of europium 153. The impurities revealed by this method may be present in only slight traces, yet by virtue of their large capture cross section for neutrons, they introduce a disturbingly large radioactivity with certainty, but a half-life of 110 days is indicated.

Two electron groups of energy 39.0 and 56.1 kev, when interpreted as K-conversion lines in gadolinium, indicate gamma-rays of energy 91.0 and 108.1 kev. Because of the interference of impurities such as europium with a six-year half-life, it is impossible to fix the periods of the gadolinium activities.

FIG. 6. Spectrograms showing the presence of terbium and europium as impurities in gadolinium.

The new information pertaining to this group of elements is assembled together in Table II.

The beta-spectrum of terbium was observed on the spectrometer by Mr. E. Salmi and that of europium by Mr. F. Shull. This investigation was made possible by the support of the Office of Naval Research.

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Microwave Determination of the Molecular Structures and Nuclear Couplings of the Methyl Halides*

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Accurate measurements on the pure rotation spectra of methyl chloride, bromide and iodide have been made with multiples of a 10 mc/sec. frequency monitored by station WWV. The measurements, with existing infra-red data, allow a complete determination of the structures of these molecules. The I_B values in g/cm$^2\times10^{-40}$ are: 63.11_{38} for $C^{12}H_3Cl^{35}$, 64.10_{09} for $C^{12}H_3Cl^{37}$, 87.68_{35} for $C^{12}H_3Br^{79}$, 88.01_{80} for $C^{12}H_3Br^{81}$, 111.8_{48} for $C^{12}H_3I^{127}$ and 117.8_{48} for $C^{13}H_3I^{127}$. The molecular dimensions obtained are: for methyl chloride, $d_{CH}=1.109$A, $d_{CCl}=1.779$A, $<$HCH $=110°\,0'$; for methyl bromide, $d_{CH}=1.104$A, $d_{CBr}=1.936$A, $<$HCH$=110°\,15'$; for methyl iodide, $d_{CH}=1.100$A, $d_{CI}=2.139$A, $<$HCH$=110°\,58'$. The nuclear quadrupole couplings, $eQ(\partial^2V/\partial z^2)$, are: for Cl^{35}, -75.13 mc/sec.; for Cl^{37}, -59.03 mc/sec.; for Br^{79}, 577.0 mc/sec.; for Br^{81}, 482.0 mc/sec.; and for I^{127}, -1934 mc/sec. Second-order effects in the hyperfine structure were observed for methyl bromide and methyl iodide.

I. INTRODUCTION

PRELIMINARY results on the microwave absorption of CH_3Cl, CH_3Br, and CH_3I have been reported.[1] In the earlier work the measurements were made with cavity wave meters and were accurate to four places only. Since that time an instrument has been constructed in our laboratory for multiplying to the millimeter region 10 mc/sec. frequencies monitored by WWV, the station of the Bureau of Standards. With it the lines of methyl chloride, bromide, and iodide occurring in the region of 8 to 11.5 mm have been measured to seven significant figures. Some improvement has also been made in the measurements of relative intensities, although we are not yet able to measure these with a high degree of accuracy.

As pointed out in the previous notes, the rotational levels of the above molecules have

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FIG. 2. Enlarged low energy spectrum of terbium (160) showing an 'N' electron group.

FIG. 6. Spectrograms showing the presence of terbium and europium as impurities in gadolinium.